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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁶ : C08G 18/32, C09D 175/00		A1	(11) International Publication Number: WO 99/07762 (43) International Publication Date: 18 February 1999 (18.02.99)												
<p>(21) International Application Number: PCT/EP98/04943</p> <p>(22) International Filing Date: 31 July 1998 (31.07.98)</p> <p>(30) Priority Data:</p> <table style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 25%;">08/906,644</td> <td style="width: 25%;">7 August 1997 (07.08.97)</td> <td style="width: 25%;">US</td> <td style="width: 25%;"></td> </tr> <tr> <td>08/906,645</td> <td>7 August 1997 (07.08.97)</td> <td>US</td> <td></td> </tr> <tr> <td>09/012,166</td> <td>22 January 1998 (22.01.98)</td> <td>US</td> <td></td> </tr> </table> <p>(71) Applicant (for all designated States except US): AKZO NOBEL N.V. [NL/NL]; Velperweg 76, NL-6824 BM Arnhem (NL).</p> <p>(72) Inventors; and</p> <p>(75) Inventors/Applicants (for US only): MOOS, Jan, Wilhelm, Ernst [NL/NL]; Pippeling 34, NL-2771 PJ Boskoop (NL). LOCHBILER, Heide, Ann [US/US]; 18680 Rosetta, East Pointe, MI 48021 (US). WEAVER, Jason, Donald [US/US]; 50041 Vinecrest, New Baltimore, MI 48047 (US). PRICE, Latoska, Nikita [US/US]; 28700 Bell Road, Southfield, MI 48034 (US). LAMB, Frances [US/US]; 16131 Winchester Drive, Northville, MI 48167 (US). RUTLEDGE, Donald, Lynn, Jr. [US/US]; 1207 Gordon Court, Clawson, MI 48017 (US). LEMAIRE, Ann, Alfred, Johanna [BE/NL]; Santhorst 49, NL-2532 KC Leiderdorp (NL). VAN ENGELEN, Antonius, Hendrikus, Gerardus [NL/US]; 2785 Quartz, Troy, MI 48098 (US). DEN BREEJEN, Catharine, Marie</p>		08/906,644	7 August 1997 (07.08.97)	US		08/906,645	7 August 1997 (07.08.97)	US		09/012,166	22 January 1998 (22.01.98)	US		<p>[NL/NL]; Van Doesburgerf 37, NL-3315 RA Dordrecht (NL).</p> <p>(74) Agent: SCHALKWIJK, Pieter, Cornelis; Akzo Nobel N.V., Patent Dept. (Dept. APTA), P.O. Box 9300, NL-6800 SB Arnhem (NL).</p> <p>(81) Designated States: AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, GH, GM, HU, ID, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).</p> <p>Published <i>With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i></p>	
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<p>(54) Title: SPRAYABLE COATING COMPOSITIONS COMPRISING OXAZOLIDINES AND ISOCYANATES</p> <p>(57) Abstract</p> <p>The present invention relates to a coating composition comprising: a) a bicyclic oxazolidine compound; b) a monocyclic oxazolidine compound; and c) an isocyanate functional compound. The invention further relates to a method of coating a substrate with the coating composition and to a substrate coated with the coating composition.</p>															

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SPRAYABLE COATING COMPOSITIONS COMPRISING OXAZOLIDINES AND ISOCYANATES

The present invention relates to a new coating composition based on
5 oxazolidines and isocyanates.

- Coatings used for painting motor vehicles and repairing the original paint are required to have good physical properties such as hardness, mechanical strength, and resistance to water, acids, and solvents. The
10 coatings are also required to have good appearance properties, which means that films must be smooth and have a high gloss and high distinctness of image (DOI). It is also desirable that all properties be retained under prolonged outdoor weathering.
- 15 For environmental reasons, it is required to use a coating composition which can be applied easily using spray application at a low volatile organic content (VOC). Coatings with a lower organic solvent content emit lower levels of solvent when they are used and so the atmosphere becomes less polluted.
20 One way to achieve a lower solvent content is to use so-called high-solids compositions. Such compositions comprise a relatively high level of non-volatile materials, such as film forming polymer, pigments and fillers, and a relatively low level of organic solvent. A problem when formulating high-solids coating compositions is that such compositions have an unacceptably high viscosity due to the high molecular weight of the conventional film forming polymer. The high viscosity gives rise to problems in spray application with poor paint atomization and poor flow-out and, consequently, low gloss levels and poor appearance.
25

The use of low-molecular weight film forming polymers, which results in adequate application viscosities, has as a disadvantage that the resulting coating is soft and marks easily. The hardness build-up of the coating is therefore unacceptable.

5

Another way to reduce the amount of volatile organic compounds in coating compositions is the use of reactive diluents. Examples of reactive diluents include aldimines and ketimines. In EP-A-0 686 654 such compounds are discussed. Other reactive diluents, such as oxazolidines, are mentioned but 10 it is stated that they have limited utility as a sole reaction partner with isocyanate because of generally slow setting film properties.

Aldimines are used as reactive diluents for low VOC paint systems. These diluents are commercially available and use of such aldimines is referred to 15 in US 5,214,086 and EP-A-0 686 654. In general these reactive diluents offer good cure and hardness development. However, they are known to cause skin irritation and to cause adhesion failure when applied in low VOC clearcoats.

20 Bicyclic oxazolidines are also used as reactive diluents for low VOC paint systems. These diluents are commercially available and use of such bicyclic oxazolidines are referred to in WO 95/14528. In general, these reactive diluents offer good durability and color stability and low toxicity. However, compositions based on bicyclic oxazolidines may exhibit an 25 unacceptably long time to cure.

Monocyclic oxazolidines are also used as reactive diluents for low VOC paint systems. Coating compositions based on monocyclic oxazolidines are also referred to in EP 0 499 188 A1. Monocyclic oxazolidines and dimers of

the same are commercially available. Coating compositions based on monocyclic oxazolidines may exhibit a loss of adhesion over time.

While the use of mono- or bi-cyclic oxazolidines has been described (see, 5 e.g., US 5,126,421), nowhere is there described the use of both monocyclic and bicyclic oxazolidines. WO 92/13907 refers to the use of monocyclic and bicyclic oxazolidines (page 7, lines 20-21), but this reference relates to the use of these compounds in the alternative and does not disclose a mixture of the two oxazolidines.

10

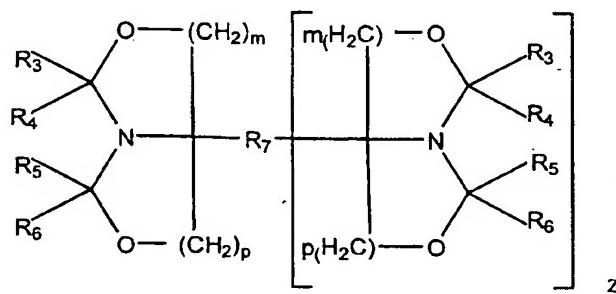
Surprisingly it was found that the use of mixtures of bicyclic and monocyclic oxazolidines in low VOC coatings provide properties that can compete with those of imines, but without loss of adhesion.

15 The present invention relates to a coating composition comprising:

- (a) a bicyclic oxazolidine compound;
- (b) a monocyclic oxazolidine compound; and
- (c) an isocyanate functional compound.

20 The variables recited in the following formulae are intended to be chosen independently both within one moiety to the next, and from one component to the next.

Preferably, the bicyclic oxazolidine component (a) has the structure:



5 wherein

z is from 0 to 9;

m and p are independently selected from 1 and 2;

R₃, R₄, R₅ and R₆ may be the same or different and are selected from the group of hydrogen, linear or branched (cyclo)alkyl, and linear or branched 10 aryl, optionally substituted;

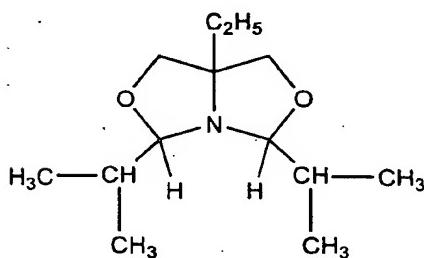
R₃ and R₄, and R₅ and R₆ may be joined together to form a 5 or 6 carbon ring with the attached carbon atom of the ring in the formula, i.e. R₃ and R₄, and R₅ and R₆ collectively represent a tetramethylene or a pentamethylene group; and

15 R₇ is a mono- or multivalent aliphatic, aromatic, arylaliphatic or cycloaliphatic moiety which may optionally contain oxygen, nitrogen, sulphur, and silica; and R₇ may be H when z=0.

Preferably, z is from 0 to 3. Preferably m and p are 1. Preferably, R₃, R₄, R₅ 20 and R₆ are selected from the group of hydrogen, phenyl, benzyl and a linear or branched C₁₋₁₂ alkyl group. More preferably, R₃, R₄, R₅, and R₆ are selected from the group of hydrogen and isopropyl.

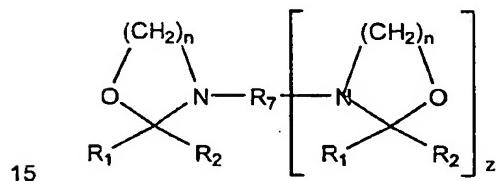
More preferably, z is 0 and R₇ is an alkyl group comprising 1 to 10 carbon atoms, more preferably methyl, ethyl or propyl. Most preferably, component (a) comprises 1-aza-3,7-dioxo-2,8-diisopropyl-5-ethyl bicyclo(3,3,0)octane, which is available commercially under the tradename "ZOLDINE® RD-20

5 from Angus Chemical Company (Buffalo Grove, IL) and has the structure:



The bicyclic oxazolidine component (a) may comprise a mixture of bicyclic
10 oxazolidines.

Preferably, the monocyclic oxazolidine (component (b)) of the present invention has the general structure:



wherein

z is from 0 to 9;

n is 2 or 3;

20 R₁ and R₂ may be the same or different and are selected from the group of hydrogen, linear or branched (cyclo)alkyl, and linear or branched aryl, optionally substituted;

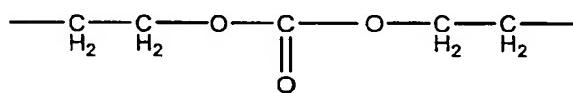
R_1 and R_2 may be joined together to form a 5 or 6 carbon ring with the attached carbon atom of the ring in the formula, i.e. R_1 and R_2 collectively represent a tetramethylene or a pentamethylene group; and

5 R_7 is a mono- or multivalent aliphatic, aromatic, arylaliphatic or cycloaliphatic moiety which may optionally contain oxygen, nitrogen, sulphur, and silica; and R_7 may be H when $z=0$.

Preferably z is from 0 to 3. Preferably n is 2. Preferably R_1 and R_2 are selected from the group of hydrogen, phenyl, benzyl and a linear or 10 branched C_{1-12} alkyl group. More preferably, R_1 and R_2 are selected from the group of hydrogen and isopropyl.

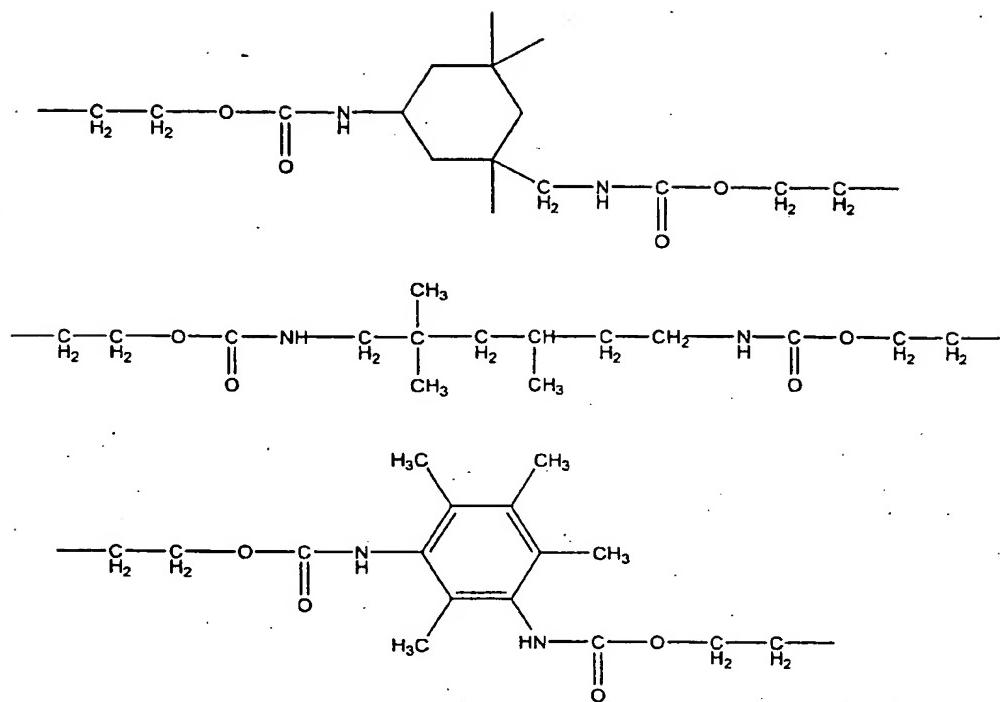
Preferably, R_7 is multivalent and more preferably R_7 is an aliphatic, 15 arylaliphatic or cycloaliphatic moiety comprising 2 to 15 carbon atoms and, optionally, ester, carbonate, and urethane groups. Monocyclic oxazolidines wherein R_7 comprises ester groups may be based on acrylate polymers such as described in GB-B-992,721.

More preferably, z is 1 and R_7 is a divalent moiety comprising either a 20 carbonate group or at least two urethane groups. Compounds having an R_7 group comprising carbonate groups are described in EP-A-0 499 188. R_7 may be for example

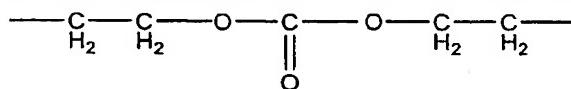


25 When R_7 comprises at least two urethane groups, R_7 may be selected from the group of the following multivalent moieties:

7



Most preferably, z is 1, n is 2, R₁ is hydrogen, R₂ is isopropyl, and R₇ is:



5. This embodiment is commercially available under the tradename "INCOZOL™" LV (from Industrial Copolymers Ltd., Preston, Lancashire, UK).

10. The monocyclic oxazolidine component (b) may comprise a mixture of monocyclic oxazolidines.

The weight ratio of bicyclic oxazolidine to monocyclic oxazolidine is preferably in the range of 5:1 to 1:2. More preferably, there is a 3:1 to 1:1

ratio of bicyclic to monocyclic oxazolidine. Most preferably, there is a 2:1 ratio.

Component (c) comprises an isocyanate functional compound and can be
5 an aromatic, aliphatic, cycloaliphatic and/or araliphatic. Component (c) can
be an isocyanurate, uretdione, biuret, allophanate, an adduct, NCO
prepolymers, or mixtures thereof.

Examples of suitable isocyanates to be used as the isocyanate functional
10 compound, or as starting materials for preparing an isocyanate functional
compound comprising an isocyanurate, biuret or uretdione structure include
organic polyisocyanates represented by the formula



wherein k is 2 or higher and R represents an organic group obtained by
15 removing the isocyanate groups from an organic polyisocyanate having
aromatically or (cyclo)aliphatically bound isocyanate groups. Preferred
diisocyanates are those represented by the above formula wherein k is 2
and R represents a divalent aliphatic hydrocarbon group having 2 to 18
carbon atoms, a divalent cycloaliphatic hydrocarbon group having 5 to 15
20 carbon atoms, a divalent araliphatic hydrocarbon group having 7 to 15
carbon atoms or a divalent aromatic hydrocarbon group having 6 to 15
carbon atoms. Examples of the organic diisocyanates which are particularly
suitable include ethylene diisocyanate, 1,3-propylene diisocyanate 1,4-
25 tetramethylene diisocyanate, 1,6-hexamethylene diisocyanate, 2,2,4-
trimethyl-1,6-hexamethylene diisocyanate, 2-methyl-1,5-diisocyanate
pentane, 2-ethyl-1,4-diisocyanate butane, 1,12-dodecamethylene
diisocyanate, cyclohexane-1,3- and -1,4-diisocyanate, 1-isocyanato-2-
isocyanatomethyl cyclopentane, IPDI, bis-(4-isocyanatocyclohexyl)-
methane, 2,4'-dicyclohexylmethane diisocyanate, 1,3- and 1,4-

bis(isocyanatomethyl)-cyclohexane, bis-(4-isocyanato-3-methylcyclohexyl)-methane, 1-methyl-2,4-diisocyanato cyclohexane, 1-isocyanato-1-methyl-4(3)-isocyanatomethyl cyclohexane, xylene diisocyanate, 1-methyl-2,4-diisocyanato benzene, $\alpha,\alpha,\alpha',\alpha'$ -tetramethyl-1,3-and -1,4-xylylene diisocyanate, 2,4- and 2,6-hexahydrotoluylene diisocyanate, 1,3- and 1,4-phenylene diisocyanate, 2,4- and 2,6-toluylene diisocyanate, 2,4- and 4,4'-diphenylmethane diisocyanate, 1,5-diisocyanato naphthalene and mixtures thereof. Aliphatic polyisocyanates containing 3 or more isocyanate groups such as 4-isocyanatomethyl-1,8-octane diisocyanate and aromatic polyisocyanate containing 3 or more isocyanate groups such as 4,4',4"-triphenylmethane triisocyanate, 1,3,5-triisocyanate benzene, polyphenyl polymethylene polyisocyanates obtained by phosgenating aniline/formaldehyde condensates, the adduct of TMP and m-tetramethylxylylene diisocyanate, and mixtures thereof may also be used. Preferred diisocyanates are 1,6-hexamethylene diisocyanate and IPDI.

Examples of suitable polyisocyanates to be used as the isocyanate functional compound or as starting materials for preparing the isocyanate functional compound comprising an allophanate structure include the above-mentioned organic polyisocyanates reacted with a mono- or polyalcohol.

Suitable mono- or polyalcohols which may be used to prepare the polyisocyanates containing allophanate groups include aliphatic, cycloaliphatic, araliphatic or aromatic mono- or polyalcohols. The mono- or polyalcohols may be linear, branched or cyclic, contain at least one carbon atom and have a molecular weight of up to 2500. The mono- or polyalcohols may optionally contain other hetero atoms in the form of, e.g.,

ether groups, ester groups, etc. However, the mono- or polyalcohols preferably do not contain hetero atoms other than the hydroxyl group(s). The molar ratio of mono- or polyalcohol to polyisocyanate is about 0.01 to 0.5, preferably about 0.04 to 0.2. Preferred mono- or polyalcohols are 5 hydrocarbon mono- or polyalcohols and mono- or polyalcohols containing ether groups. The hydrocarbon mono- or polyalcohols preferably contain 1 to 36, more preferably 1 to 20 and most preferably 1 to 8 carbon atoms.

Examples of suitable monoalcohols include methanol, ethanol, n-propanol, 10 isopropanol, n-butanol, isobutanol, t-butanol, n-pentanol, 2-hydroxy pentane, 3-hydroxy pentane, the isomeric methyl butyl alcohols, the isomeric dimethyl propyl alcohols, neopentyl alcohol, n-hexanol, n-heptanol, n-octanol, n-nonanol, 2-ethyl hexanol, trimethyl hexanol, decanol, dodecanol, tetradecanol, hexadecanol, octadecanol, 2,6,8- 15 trimethylnonanol, 2-t-butyl-cyclohexanol, 4-cyclohexyl-1-butanol, cyclohexanol, benzyl alcohol, phenol, the cresols, the xylenols, the trimethylphenols, 2,4,6-trimethyl benzyl alcohol, branched chain primary alcohols and mixtures thereof (which are available from Henkel (Minneapolis, MN) under the "STANDAMUL®" trademark) and mixtures of 20 linear primary alcohols (which are available from Shell (Houston, TX) under the "NEODOL®" trademark).

Preferred ether-containing monoalcohols include ethoxy methanol, methoxy ethanol, ethoxy ethanol, the isomeric methoxy or ethoxy propanols, the isomeric propoxy methanols and ethanols, the isomeric methoxy butanols, the isomeric butoxy methanols, furfuralcohol and other monoalcohols which have a molecular weight of up to 2500 and are based 25 on ethylene oxide, propylene oxide and/or butylene oxide. It is also

possible in accordance with the present invention to use mixtures of the previously described monoalcohols.

Examples of suitable polyalcohols having two or more hydroxyl groups include ethane diol, 1,3-propanediol, 1,2-propanediol, 2-methyl-1,3-propanediol, 1,4-butanediol, 1,3-butanediol, 1,6-hexanediol, neopentylglycol, glycerol, pentaerythritol, trimethylol propane, ditrimethylol propane, 1,4-cyclohexane dimethanol, the monoester of neopentylglycol and hydroxy pivalic acid, 2,2,4-trimethyl pentanediol, and dimethylol propionic acid, and mixtures thereof. Other preferred polyalcohols for the production of suitable polyurethanes include polyester and polyether diols having a number average molecular weight of less than 1000, for example the polyester diol prepared from 1 mole of phthalic anhydride and 2 moles of neopentyl glycol. It is also possible in accordance with the present invention to use mixtures of the polyalcohols and mixtures of a polyalcohol and the previously described monoalcohols.

Preferably, isocyanate functional compound comprising an allophanate structure are prepared from 1,6-hexamethylene diisocyanate and/or IPDI reacted with an alcohol, preferably butanol.

Polyisocyanate adducts include the adduct of TMP and m-tetramethylxylylene diisocyanate.

The NCO prepolymers are prepared from the previously described monomeric polyisocyanates, preferably monomeric diisocyanates, and organic compounds containing at least two isocyanate-reactive groups, preferably at least two hydroxy groups. These organic compounds include high molecular weight compounds having number average molecular

weights of 400 to about 6,000, preferably 800 to about 3,000, and optionally low molecular weight compounds with molecular weights below 400. Products obtained by reacting polyisocyanates exclusively with low molecular weight compounds are polyisocyanate adducts containing 5 urethane groups and are not considered to be NCO prepolymers. Examples of the high molecular weight compounds are polyester polyols, polyether polyols, polyhydroxy polycarbonates, polyhydroxy polyacetals, polyhydroxy polyacrylates, polyhydroxy polyester amides and polyhydroxy polythioethers. The polyester polyols, polyether polyols and polyhydroxy 10 polycarbonates are preferred. Further details concerning the low molecular weight compounds and the starting materials and methods for preparing the high molecular weight polyhydroxy compounds are disclosed in US 4,701,480. These NCO prepolymers are prepared in known manner by the reaction of the above-mentioned starting materials at an NCO/OH 15 equivalent ratio of about 1.05:1 to 10:1 preferably about 1.1:1 to 3:1. This reaction may take place in a suitable solvent which may optionally be removed by distillation after the reaction along with any unreacted volatile starting polyisocyanates still present.

20 Preferred aromatic isocyanates are "DESMODUR®" IL (isocyanurate of toluene diisocyanate (TDI)), "DESMODUR®" VL (diphenyl methyl-4,4'-diisocyanate prepolymer) and "DESMODUR®" L75 (TDI(trimethylol propane(TMP) adduct) (all available from Bayer, Pittsburgh, PA). Preferred aliphatic or cycloaliphatic isocyanates are "DESMODUR®" N3400 25 (uretdione of hexamethylene diisocyanate (HDI)), "DESMODUR®" N3300 (isocyanurate of HDI) and "VESTANAT®" T1890 (isocyanurate of isophorone diisocyanate (IPDI))(available from Hüls, Chicago, IL). Preferably, component (c) is an aliphatic or cyclo-aliphatic isocyanate. More preferably, component (c) is mixture of isocyanate functional

compounds, preferably with at least one uretdione and at least one isocyanurate. More preferably, the mixture comprises "DESMODUR®" N3400, "DESMODUR®" N 3300 and "VESTANAT®" T1890.

- 5 The isocyanate functional compound comprising a uretdione structure is preferably present in an amount of at least 5% by weight, preferably at least 10% by weight, based on the solids content of the polyisocyanate component (c).
- 10 Preferably, the coating composition of the present invention further comprises a component (d). Component (d) comprises a resin comprising a functional group selected from hydroxyl and amine. The hydroxyl functional resin may be an acrylic, polyester, polyether, polyurethane functional resin or other constituent materials known in the art, and may be a mixture 15 thereof. Some commercial examples of acrylic hydroxyls are "JONCRYL®" 500 (S.C. Johnson & Son, Inc., Racine, WI) and AU608 (Rohm and Haas, Philadelphia, PA). Preferably, the hydroxyl functional resin is a mixture of a polyester resin and a polyurethane polyol.
- 20 Alternately, component (d) comprises an amine functional resin. Suitable compounds may be aliphatic, aromatic, cycloaliphatic and/or araliphatic, may contain a saturated, an unsaturated group, O, S or N, and include ethylene diamine, ethylene glycol diamine, propylene glycol diamine, and cycloaliphatic diamines. Preferably, the amine functional resin is an 25 aspartic acid ester. Suitable resins are available commercially from Bayer under the tradenames "DESMOPHEN®" XP 7052 (sterically hindered amines - adduct of 2 moles diethylmaleate with the amine "LAROMIN®" C 260 (BASF, Germany)) and "DESMOPHEN®" XP 7053 (adduct of 2 moles diethylmaleate with the amine PACM 20).

Preferably, component (d) is a hydroxyl functional resin.

Preferably, the percentages by weight of components (a) to (d) on total
5 vehicle solids are: (a) 2-40%, (b) 3-20%, (c) 30-70%, and (d) 5-30%; more
preferably, (a) 8-30%, (b) 4-15%, (c) 35-65% and (d) 7-28%; and most
preferably (a) 15-25%, (b) 5-13%, (c) 40-60% and (d) 10-25%.

The coating composition of the present invention may also comprise (e)
10 hydroxyl or other functional reactive diluents, which may lower viscosity,
lower VOC and boost reactivity.

The coating composition of the present invention may also comprise
additional components such as solvents, catalysts, stabilizers, fillers,
15 rheology control agents, flow additives, leveling additives, dispersing
agents and other components known to persons skilled in the art. Suitable
solvents include methyl amyl ketone, butyl acetate, amyl acetate, ethoxy
ethyl propionate and xylene. Suitable catalysts include aromatic or aliphatic
carboxylic acids, arylsulphonic acids and organometal compounds. Acids
20 which are useful are formic acid, acetic acid, mono-, di-, and trichloro acetic
acid, oxalic acid, maleic acid, malonic acid, fumaric acid, heptanoic acid,
pelargonic acid, isononanoic acid, benzoic acid, 4-hydroxybenzoic acid,
mono-, di-, and trichlorobenzoic acid, and salicylic acid and anhydrides
thereof. Preferred acids are acetic acid, heptanoic acid, and benzoic acid.
25 Useful organometal compounds include zinc alkanoate, such as zinc
octoate, dibutyltin dilaurate, dibutyltin (bis)mercaptide, dibutyltin diacetate,
and dibutyltin sulphide. Preferred is an organo-tin catalyst such as dibutyltin
dilaurate. More preferably, mixtures of organo-tin and organo-zinc catalysts
are used. Also mixtures of the above-mentioned catalysts may be used.

- Optionally pigments may be present in the coating composition of the present invention. Useful pigments are various types common to the art which include but are not limited to titanium dioxide, graphite, carbon black,
- 5 zinc oxide, calcium sulphide, chromium oxide, zinc sulphide, zinc chromate, strontium chromate, barium chromate, lead chromate, lead cyanamide, lead silico chromate, yellow nickel titanium, yellow chromium titanium, red iron oxide, yellow iron oxide, black iron oxide, naphtol red and browns, anthraquinones, dioxa zinc violet, isoindoline yellow, arylide yellow and
- 10 oranges, ultramarine blue, phthalocyanine complexes, amaranth, quinacridones, halogenated thioindigo pigments, extender pigments such as magnesium silicate, aluminium silicate, calcium silicate, calcium carbonate, fumed silica, barium sulfate, and zinc phosphate.
- 15 Preferably, the coating composition comprises less than 500 g/l (4.2 lb/gal) of volatile organic solvent based on the total composition, more preferably less than 480 g/l (4.0 lb/gal), most preferably less than 420 g/l (3.5 lb/gal). The solid resin content preferably is higher than 50%, more preferably higher than 52%, most preferably higher than 58%.
- 20 Coating compositions of the present invention are useful as clearcoats, basecoats, topcoats and primers.
- The coating composition of the present invention can be used in the
- 25 preparation of coated substrates. These substrates include glass, ceramics, paper, wood, plastic and metal.
- The coating composition is especially useful in the refinish industry, in particular the body shop, to repair automobiles. The coating composition is

- also applicable in the automotive industry for the finishing of large transport vehicles, such as trains and buses, and can also be used in airplanes. The substrate may be uncoated material or can be primed. The substrate may also be coated with paint products applied at the time of manufacture or 5 just prior to application of the compositions of the present invention. The coating composition can be applied using conventional spray equipment or high volume low-pressure spray equipment resulting in a high quality finish. Other modes of application are roller coating, brushing, sprinkling, flow coating, dipping, electrostatic spraying or electrophoresis, with spraying 10 being preferred. Exemplary metal substrates include steel, aluminium, copper, zinc, magnesium, and alloys thereof. Curing temperatures are preferably between 0 and 80°C, and more preferably between 20 and 60°C.
- 15 The following examples illustrate the preparation of compositions according to the present invention. The examples are illustrative of the invention and contain a best mode. Comparative examples are included to illustrate the benefits of the present invention over other systems.

Examples

Example 1

A clearcoat according to the present invention was prepared as follows.

- 5 Percentages given are by weight of each component mixture. The components are listed in order of addition. All components were added while stirring and were mixed on an air mixer for a minimum of 20 minutes after the addition was complete.

	<u>Component A</u>	<u>Amount (%)</u>	<u>Type</u>
10	PE	42.97	Binder (see below)(80.5% solids)
	PUPO	13.55	Binder (see below)(60.8% solids)
	Byk 358	0.23	Flow & Lev.agent (Byk Chemicals, Wallingford, CT)
15	Byk 333	0.28	Flow & Lev. agent(Byk Chemicals)
	Fascat 4202	0.10	Tin Catalyst (Elf Atochem, Philadelphia, PA)
	Nuxtra Zinc	0.65	Zinc Catalyst (Hüls)
	butyl acetate	13.81	solvent (Eastman Chemical, Kingsport, TN)
20	EXXATE 600	5.35	solvent (mixture of esters)(Exxon, Houston, TX)
	methyl isobutyl ketone	6.42	solvent (Eastman Chemical)
	butyl cellosolve acetate	2.14	solvent
25	dibasic ester aliphatic	1.66	solvent (DuPont, Wilmington, DE)
	methyl amyl ketone	12.87	solvent (Eastman Chemical)

Preparation of the polyester resin (PE):

Into a 10 liter round bottom reaction vessel equipped with a stirrer, packed column, condenser, heating mantle, thermometer, and nitrogen inlet, were charged:

- 5 2608 g of hexahydrophthalic anhydride,
- 2981 g of trimethylol propane,
- 2015 g of isononanoic acid,
- 195 g of phthalic anhydride, and
- 9.2 g of an aqueous solution comprising 85% phosphoric acid.
- 10 The reaction mixture was heated under a nitrogen stream of 10 liter/hour. The temperature of the mixture was gradually raised to 240°C. The reaction water was distilled off at a rate such that the temperature at the top of the packed column did not exceed 103°C. After the reaction was run at 240°C for one hour, the nitrogen stream was increased to 50 liter/hour and the
- 15 reaction continued at 240°C until an acid value of 9.3 was reached. The reaction mixture was then cooled to 130°C and diluted with 1,817 g of butylacetate.

- 20 A polyester polyol solution was obtained having a solids content of 80.5%, a viscosity of 7.5 Pa.s measured at 20°C, an acid value of 9.3 and a hydroxyl value of 145, both based on solids. The polyester polyol had a Tg of -2°C. The polyester polyol had an Mn of 1,900 and an Mw of 4,500 (as measured by gel permeation chromatography using polystyrene as a standard).

25

Preparation of the polyurethane polyol (PUPO):

Into a two liter round bottom reaction vessel equipped with a stirrer, packed column, condenser, heating mantle, thermometer, and nitrogen inlet, were charged:

426 g of 2-butyl-2-ethyl-1,3-propanediol,
 440 g of butyl acetate, and
 0.11 g of dibutyltin dilaurate.

The mixture was heated under a nitrogen atmosphere to 55°C. Then 934 g
 5 of a 70% solution in butylacetate of the isocyanurate trimer of IPDI
 (available as "VESTANAT®" T 1890 E from Hüls) were added over a period
 of about 60 minutes. During this addition the temperature was not allowed
 to exceed 70°C. After complete addition, the mixture was post-reacted at
 10 70°C until the NCO-content, calculated on solid ingredients, dropped below
 0.1 wt%. After cooling, a clear polyurethane polyol solution was obtained
 having a viscosity (at 20°C) of 2.8 Pa.s, a solids content of 60.8%
 (determined at 150°C for 60 minutes) and a hydroxyl value of about 136
 calculated on solids. Using polystyrene as a standard, gel permeation
 chromatography (GPC) indicated an Mn of 1,300 and a Mw 1,800. The
 15 polyurethane polyol had a Tg of 81°C.

<u>Component B</u>	<u>Amount (%)</u>	<u>Type</u>
DESMODUR N3390	53.73	HDI isocyanurate (Bayer) (90% solids in butylacetate)
VESTANAT T1890E	34.51	IPDI isocyanurate (Hüls) (70% solids in butylacetate)
DESMODUR N3400	10.89	HDI Uretidine (Bayer)

25	<u>Component C</u>	<u>Amount (%)</u>	<u>Type</u>
ZOLDINE RD20 LC	66.00	Bicyclic oxazolidine (Angus Chemical)	
INCOZOI LV	34.00	Monocyclic oxazolidine (Industrial Copolymers)	

Components A, B and C were mixed by weight as follows:

40.00 grams A

91.00 grams B

41.81 grams C

- 5 To this mixture, 15.20 grams of butyl acetate were added in order to bring the DIN Cup 4 viscosity below 20 seconds. (Viscosity was measured in a DIN flow cup number 4 in accordance with DIN 53221-1987.) The VOC of the composition was 2.38 lb/gal (285 g/l). (VOC was measured in accordance with ASTM 3960-92.)

10

Comparative Example (1A)

- For comparative purposes, a clearcoat was made using the same components as in Example 1 except that instead of using the oxazolidine mixture C of Example 1, the aldimine "VESTAMIN®" A-139 (available from Hüls) was used. The VOC of the composition was 2.30 lb/gal (276 g/l).

Experimental Procedure.

Two E-coated cold rolled steel panels were prepared as follows:

- The steel panels were degreased with M600 Degreaser (commercially available from Akzo Nobel Coatings Inc., Norcross GA) and scuffed with a red Scotch Brite pad. Then the panels were degreased again with M600. "AUTOBASE" (commercially available from Akzo Nobel Coatings) was reduced with 790 Reducer Slow (commercially available from Akzo Nobel Coatings) at a 100:80 mixing ratio (by volume). The following AUTOBASE color formula was used:

	<u>wt.% (before addition of reducer)</u>
toner 777	10
toner 666	10
toner 333C	40

toner 956 40

After reduction with 790 Reducer, three coats plus one drop coat of basecoat was applied using a SATA NR95 HVLP spray gun (with 1.3 tip set) at 10 psi air pressure at the gun at 70°F and 16% relative humidity. A 5 5-10 min. flash was given between coats and 30 minutes before application of the clearcoat.

The compositions of Example 1 and Comparative Example 1A were applied in two coats by means of HVLP spraying (SATA NR95 with 1.5 tip 10 set at 10 psi) at 70°F and 16% relative humidity. The first coats were allowed to flash for 10 minutes before application of the second coat. All clearcoats were applied to a dry filmbuild of approximately 3 mils. All clearcoats were baked for 1 hour at 140°F. They were then tested for cure speed. Hardness and adhesion were tested after 1 day storage at 70°F and 15 16% RH. The results are shown in Table I:

TABLE 1

	Ex. 1	Comp.Ex. 1A
<u>Cure [1]</u> (60 min. @ 60 °C)	Cured, slightly tacky	Cured
<u>Persoz Hardness [2]</u> after 1 day @ at 70 °F, 16% RH	83	209
<u>Adhesion [3]</u> after 1 day @ 70 °F, 16% RH	9-10	6-7

[1] Cure determined manually by pressing a finger in the fresh clearcoat film.

[2] Persoz Hardness according to ISO 1522-1973 using steel instead of glass plate.

20 [3] Cross Cut adhesion testing with tape, "10" indicating no adhesion failure, "0" indicating complete adhesion failure.

As can be seen from the above data, the composition of Example 1, which embodies the present invention, showed good adhesion, acceptable hardness and an acceptable time to cure. In comparison, the composition that contained the aldimine instead of the oxazolidines (Comparative 5 Example 1A) showed good cure and hardness level, but shows poor adhesion.

Example 2

10 A primer was made according to the present invention and prepared from the following: (Percentages given are by weight of the total coating composition mixture).

4.0%	DESMOPHEN 670-80 polyester resin (Bayer)
4.0%	JONCRYL SCX 910 acrylic resin (S.C. Johnson)
12.8%	ZOLDINE RD -20
4.2 %	INCOZOL LV
14 %	DESMODUR IL, an aromatic polyisocyanate based on TDI (51% solids in butylacetate)(Bayer)
15.8 %	MONDUR MRS, an aromatic polyisocyanate based on diphenylmethane 4,4 diisocyanate (Bayer)
3%	DESMODUR 3400, uretdione of HDI (Bayer)

Other components (42.3%)

20.8%	Pigments, including anti-corrosive pigments, fillers, TiO ₂ and Carbon black
4.7 %	Flow agent, polyacrylate type
16.8 %	Solvents, including esters and ketones

The primer was prepared as follows. Into a clean, dry tank were charged and mixed:

776.7115 g DISPERBYK-110 (dispersant)(Byk)
268.0569 g butyl acetate

5 While mixing, the following were added:

650.6976 g DESMOPHEN 670A-80
650.6976 g JONCRYL SCX 910

The following were then added under mixing:

53.4307 g COSMIC BLACK D-2 (carbon black)
10 (Ebonex, Melvindale, MI)
379.9135 g BLANC FIXE MICRO (filler pigment)
(Sachtleben Chemie, Duisburg, Germany)
975.1328 g TALC 399 LO MICRON MAGNESIUM SILICATE
(filler pigment) (Barretts Minerals, Bethlehem, PA)
15 703.8150 g WOLLASTOCOAT 10ES (filler pigment)
(Nyco, Willsboro, NY)
703.8755 g TI-PURE R-902 28 (TiO₂) (DuPont, Wilmington, DE)
626.4065 g HALOX CW 491 (anticorrosive pigment)
(Halox Corp., Hammond, IN)

20 Mixing was continued for five minutes, then DYNOMILL ground to 20-25 microns, as verified by a quality control grind check. The following was then added while mixing:

600.0000 g butyl acetate

A quality control check was made of 1 quart to check the percentage of 25 non-volatiles (acceptable is 81.8 ± 2.0). Also checked was Stormer viscosity (95-100 Krebs units) and density (14.07 ± 0.5 lbs./gal). It is important that the tank be kept closed during this quality control check. The batch must be filled within 24 hours. The batch was filtered through a 100 micron bag.

Activator

Into a clean, dry tank were charged:

200.7000 g ZOLDINE RD-20

5 66.6700 g INCOZOL LV

This was mixed on low speed for 5 minutes. Then the following was added and mixed together for 30 minutes:

1.4400 g DESMOPHEN 670A-80

0.3600 g MULTIFLOW (flow and leveling agent)

10 (Monsanto, St. Louis, MO)

37.5000 g methyl amyl ketone

37.5000 g ethyl amyl ketone

88.8900 g butyl acetate

A 1 quart sample was subject to a quality control check for density (7.42 ± 0.5) and viscosity in a # 4 cup at 77°F (12.0 ± 2.0). It is important that the tank be kept closed during the quality control check.

Hardener

Under conditions to minimize moisture content, into a clean, dry, closed tank blanketed with nitrogen, were dosed and mixed on low speed for 10 minutes:

25 4.5359 lb DESMODUR IL

5.1454 lb MONDUR MRS

0.9681 lb DESMODUR 3400

Water content was checked on a 4 oz. sample. Water content should be less than 1500 ppm. Then was added:

0.1817 lb n-butyl acetate

0.0872 lb DOWANOL PMA (Dow, Midland, MI)

0.0654 lb butyl cellosolve acetate

0.0291 lb Aromatic 100 (Exxon)

Water content was again checked. The solvent blend was mixed on low speed for 30 minutes. A 1 quart sample was subject to a quality control check for density (9.65 ± 0.5), percentage non-volatiles (75.1 ± 2.0) and viscosity with #4 cup at 77°F (60.0 ± 3.0). It is important that the tank be kept closed during the quality control check. The batch must be filled within 24 hours and filtered through a 10 micron bag. The VOC of the composition was 2.8 lb/gal. (335 g/l)

10 Comparative Examples

For comparative purposes, three additional primers were made using the same components as in Example 2 except that instead of using the oxazolidine mixture of Example 2 the following were used:

15 Comparative Example 2A was made with a di-monocyclic oxazolidine ("INCOZOLTM" LV) as the sole oxazolidine component.

Comparative Example 2B was made with a bicyclic oxazolidine ("ZOLDINE[®]" RD 20) as the sole oxazolidine component.

Comparative Example 2C was made with an aldimine ("VESTAMIN[®]" A139 available from Hüls) instead of the oxazolidine component.

20 The VOC of these compositions was 2.8 lb/gal (335 g/l).

The composition of Examples 2 and Comparative examples 2A, 2B, and 2C were applied using a SATA 95 HVLP spray gun and both ambient air cured and baked 60 minutes at 60°C (140°F). They were tested for dry times (dust free and tack free) and crosshatch adhesion. Example 2' is the same as example 2 except that the dust free and tack free times were measured under 25 % relative humidity versus 70% relative humidity of Example 2. Comparative Examples 2A, 2B and 2C were subject to 21% relative humidity. The results are shown below in Table 2.

TABLE 2

Ex.	Viscosity (#4 Ford, 25°C, sec.)	Dust Free (min.)	Tack Free (min.)	Pot life (min.)	Adhesion (X-Cut)	Temp/ Relative Humidity
2	17	27	28	50	10	71.8°F/70%
2'	17	46	65	50	10	71°F/25%
2A	18	50	100	40	10	71°F/21 %
2B	17	100	100	45	10	71°F/21 %
2C	22	9	19	30	10	71°F/21 %

Pot life is the time between the initial mixing of all components and the point where the viscosity has increased to 2 times the initial viscosity.

5

Example 2 (mixture of oxazolidines) showed improved potlife over 2C (aldimine), and to a lesser extent over 2A (monocyclic alone) and 2B (bicyclic alone). The tack free time of example 2 is much improved over both the 2A (monocyclic) and 2B (bicyclic), and dust free time is much 10 improved over 2B (bicyclic). Viscosity is lower in example 2 over 2C (aldimine). Adhesion was equivalent throughout.

Example 3

15 A clearcoat according to the present invention (3C) and four comparative examples (3A, 3B 3D and 3E) were prepared as in Example 1 using the following ingredients.

Clear Formulation

PE	42.75 g
PUPO	13.48 g
20% BYK 310 in BuAc (flow and lev. Agent) (Byk)	2.98 g
10% T-12 in BuAc	99 g
methyl N-amyl ketone	9.95 g
methyl N propyl ketone	29.85 g
Total	100 g
VOC	4.45 Lbs/Gal

Hardener Formulation

DESMODUR N-3390	92.27 g
methyl N-propyl ketone	7.73 g
Total	100 g
VOC	1.55 Lbs/Gal

5 Activator Formulas

	A	B	C	D
ZOLDINE RD-20	100g	-	66g	-
INCOZOL LV	-	100g	34g	-
VESTAMIN A-139	-	-	-	100g
Total	100g	100g	100g	100g
VOC	0.00 Lbs/Gal	0.00 Lbs/Gal	0.00 Lbs/Gal	0.00 Lbs/Gal

Example 3C and comparative examples

	3A	3B	3C	3D	3E
Clear	24.39	24.39	24.39	24.39	-
Hardener	48.78	48.78	48.78	48.78	-
Activator A	24.39	-	-	-	-
Activator B	-	24.39	-	-	-
Activator C	-	-	24.39	-	-
Activator D	-	-	-	24.39	-
methyl N-amyl ketone	2.44	2.44	2.44	2.44	-
AUTOCLEAR II	-	-	-	-	100 ml
AUTOCRYL Hardener	-	-	-	-	50 ml
AUTOCRYL Reducer Medium	-	-	-	-	30 ml
Total	100g	100g	100g	100g	180 ml
VOC (Lbs/Gal)	2.1	2.1	2.1	2.1	4.4
X-Link	87%	70%	80%	103%	-

Conditions: 38% rH, 80°F

- Example 3C was prepared according to the present invention using a mixture of bicyclic and monocyclic oxazolidines. Comparative Example 3A was prepared using only bicyclic oxazolidine. Comparative Example 3B was prepared using only monocyclic oxazolidine. Comparative Example 3D was prepared using aldimine instead of oxazolidine. Comparative Example 3E is used to show a conventional composition (using "AUTOCLEAR®" and "AUTOCRYL®" components available from Akzo Nobel Coatings Inc.) with acceptable properties that has a higher VOC content than the composition of the present invention.

Results

Test results are shown in the following tables comparing potlife, drying characteristics, physical properties (adhesion and Persoz hardness), gloss and DOI (distinctness of image).

5

Potlife

	Initial	30 min	60 min	Potlife
3A	17.87 sec	19.04 sec	19.85 sec	> 60 minutes
3B	19.32 sec	21.12 sec	23.00 sec	> 60 minutes
3C	18.31 sec	23.47 sec	26.97 sec	60 minutes
3D	21.29 sec	34.28 sec	48.22 sec	30 minutes
3E	16.12 sec	-	17.56 sec	> 60 minutes

The results show that the composition according to the present invention, 3C, shows an improved potlife over 3D (the composition using the aldimine instead of the oxazolidine). The potlife of 3D is short, but may still be acceptable for small applications such as spot and panel repairs. However, it would be unacceptable for a complete overspray.

Drying Characteristics - Bake 60°C

	Dry to Handle
3A	>120min NOK
3B	25 min excellent
3C	35 min good
3D	15 min outstanding
3E	25 min excellent

15 NOK = not OK

Drying characteristics of the bi-/mono-cyclic oxazolidine mixture 3C were improved over the bicyclic oxazolidine alone (3A) at 60°C. In fact, 3A was still wet after the normal 35 minute bake period at 60°C. This was not acceptable.

5

Physical Properties

	Adhesion			Persoz hardness		
	3 Day	6 Day	10 Day	3 Day	6 Day	10 Day
3A	10	10	10	209/NOK*	244/NOK*	255/NOK*
3B	2	1	1	51	57	68
3C	10	10	10	103	119	133
3D	5	5	5	74	80	85
3E	10	10	10	116	142	158

NOK = not OK

* panels of 3A had to baked for > 120 minutes in order to cure to dry to handle. The panels were deemed not acceptable because the hardness would not have been acceptable after 10 a normal 35 minute cure.

The baked panels were tested for adhesion and Persoz hardness. The composition of the present invention comprising the bicyclic and monocyclic oxazolidine (3C) shows improved adhesion and hardness over 15 3B (monocyclic oxazolidine alone) and over 3D (aldimine), and also improved hardness over 3A (bicyclic oxazolidine alone) under normal baking conditions (see note above).

	Gloss			DOI		
	3 Day	6 Day	10 day	3 Day	6 Day	10 Day
3A	82.5	82.9	83.4	80	80	80
3B	85.0	82.4	82.1	85	85	85
3C	83.3	81.8	78.9	80	80	80
3D	82.9	83.7	83.1	80	80	80
3E	81.9	80.0	79.8	80	80	80

Gloss is measured in accordance with ISO 2813:1994 (angle 20°) and is expressed in GU.

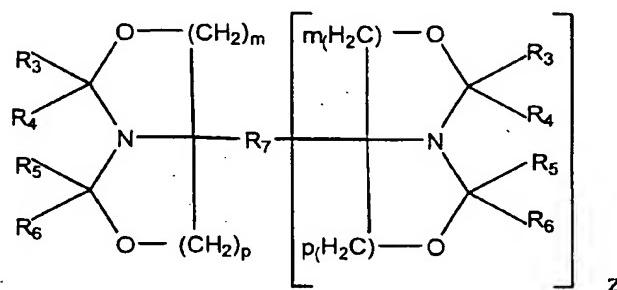
- 5 DOI is determined by projecting a standard (original) image on a coating. The sharpness of the reflected image and of the original image is compared. If the reflected image is (nearly) as sharp as the original image, the DOI of the coating is high. If the reflected image is much less sharp than the original image, the DOI of the coating is low.
- 10 The gloss and DOI (distinctness of image) data indicate that appearance of all of the panels is acceptable. No appreciable difference was seen after 6 and 10 days.
- One skilled in the art will appreciate that the present invention can be practiced by other than the described embodiments, which are presented
- 15 for purposes of illustration and not of limitation, and the present invention is limited only by the claims that follow.

We claim:

1. A coating composition comprising:

(a) a bicyclic oxazolidine compound of the structure:

5



wherein

z is from 0 to 9;

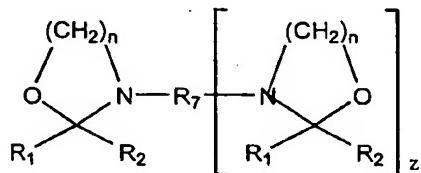
10 m and p are independently selected from 1 and 2;

R₃, R₄, R₅ and R₆ may be the same or different and are selected from the group of hydrogen, linear or branched (cyclo)alkyl, and linear or branched aryl, optionally substituted;

15 R₃ and R₄, and R₅ and R₆ may be joined together to form a 5 or 6 carbon ring with the attached carbon atom of the ring in the formula, i.e. R₃ and R₄, and R₅ and R₆ collectively represent a tetramethylene or a pentamethylene group; and

20 R₇ is a mono- or multivalent aliphatic, aromatic, arylaliphatic or cycloaliphatic moiety which may optionally contain oxygen, nitrogen, sulphur, and silica; and R₇ may be H when z=0;

(b) a monocyclic oxazolidine compound of the structure:



5 wherein

z is from 0 to 9;

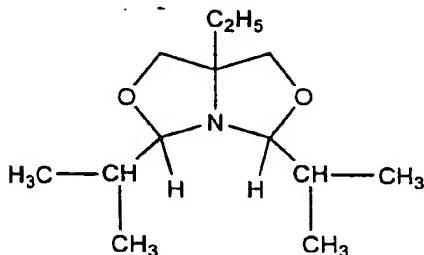
n is 2 or 3;

10 R_1 and R_2 may be the same or different and are selected from the group of hydrogen, linear or branched (cyclo)alkyl, and linear or branched aryl, optionally substituted;

15 R_1 and R_2 may be joined together to form a 5 or 6 carbon ring with the attached carbon atom of the ring in the formula, i.e. R_1 and R_2 collectively represent a tetramethylene or a pentamethylene group; and R_7 is a mono- or multivalent aliphatic, aromatic, arylaliphatic or cycloaliphatic moiety which may optionally contain oxygen, nitrogen, sulphur, and silica; and R_7 may be H when $z=0$; and

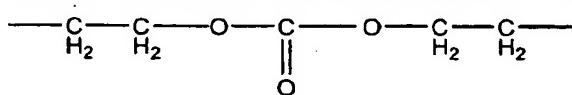
(c) an isocyanate functional compound.

2. The composition of claim 1 wherein component (a) has the formula:



3. The composition according to any of the preceding claims wherein in component (b) $z=1$.

4. The composition according to claim 3 wherein in component (b) $z=1$.



10

5. The composition according to any of the preceding claims wherein component (c) is selected from an aromatic and an aliphatic isocyanate functional compound.
 - 15 6. The composition according to any of the preceding claims further comprising (d) a resin comprising a functional group selected from hydroxyl and amine.
 7. The composition of claim 6 wherein component (d) comprises a hydroxyl functional resin.
 - 20 8. The composition according to any of the preceding claims further comprising (e) a hydroxy functional reactive diluent.

9. A method of coating a substrate comprising coating the substrate with the coating composition according to any of the preceding claims.
- 5 10. A coated substrate coated with the composition according to any of the preceding claims .

INTERNATIONAL SEARCH REPORT

Internatinal Application No
PCT/EP 98/04943

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 C08G18/32 C09D175/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 C08G C09D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 96 08308 A (ANGUS CHEMICAL COMPANY) 21 March 1996 see page 10, line 1 - line 3; claims 51,53,56,58,59 ---	1,2,5-7, 9,10
A	US 5 157 100 A (J.R.BABJAK ET AL.) 20 October 1992 see claims 9,11; examples 8-11 ---	1,2,5-10
A	US 3 864 335 A (W.D.EMMONS) 4 February 1975 see column 5, line 3 - line 9; claims 1,14; example 4 ---	1,3-5,9, 10 -/-

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

* Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
- "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search

Date of mailing of the international search report

30 November 1998

04/12/1998

Name and mailing address of the ISA

European Patent Office, P.O. Box 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
Fax: (+31-70) 340-3016

Authorized officer

Angiolini, D

INTERNATIONAL SEARCH REPORT

Inter	inal Application No
PCT/EP 98/04943	

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5 219 979 A (A.GRECO) 15 June 1993 see column 7, line 53 - line 65; claim 1; example 1 & EP 0 499 188 A cited in the application -----	1,3-7
A		

INTERNATIONAL SEARCH REPORT

Information on patent family members

Inte	rnational Application No
	PCT/EP 98/04943

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